

Nonlinear, ground-state, pump-probe spectroscopy

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A theory of pump-probe spectroscopy is developed in which optical fields drive two-quantum, Raman-like transitions between ground state sublevels. Three fields are incident on an ensemble of atoms. Two of the fields act as the pump field for the two-quantum transitions. The absorption or gain of an additional probe field is monitored as a function of its detuning from one of the fields which constitutes the pump field. Although the probe absorption spectrum displays features common to those found in pump-probe spectroscopy of single-quantum transitions, new interference effects are found to modify the spectrum. Many of these features can be explained within the context of a dressed atom picture.

I. INTRODUCTION

Of fundamental interest in nonlinear spectroscopy is the response of an atomic vapor to the simultaneous application of a pump and a probe field. A calculation of the probe field absorption is relatively straightforward [1,2] in the weak probe field limit. Let Ω and Ω' denote the pump and probe field frequencies, $\Delta = \Omega - \omega$ the pump field detuning from atomic resonance ω , and $\delta_1 = \Omega' - \Omega$ the probe-pump detuning. For a pump field detuning $|\Delta| \gg \gamma_e, \chi$, where γ_e is the upper state decay rate and χ is a pump-field Rabi frequency, one finds the spectrum to consist of three components. There is an absorption peak centered near $\delta_1 = -\Delta$ ($\Omega' = \omega$), an emission peak centered near $\delta_1 = \Delta$ ($\Omega' = 2\Omega - \omega$) and a dispersive like structure centered near $\delta_1 = 0$. Experimentally, a spectrum exhibiting all these features was first obtained by Wu *et al.* [3]. The absorption and emission peaks can be given a simple interpretation in a dressed-atom picture [4], but the non-secular structure centered at $\delta_1 = 0$ is somewhat more difficult to interpret [5,6]. The width of these spectral components is on the order of γ_e , neglecting any Doppler broadening.

The spectral response can change dramatically when atomic recoil accompanying the absorption or emission of radiation becomes a factor [7], as in the case of a highly collimated atomic beam or for atoms cooled below the recoil limit. In this limit, the absorption and emission peaks are each replaced by an absorption-emission doublet, and the dispersive-like structure is replaced by a pair of absorption-emission doublets. The spectrum can be given a simple interpretation in terms of a dressed atom theory, including quantization of the atoms' center-of-mass motion [7]. It turns out, however, that at most one absorption-emission doublet (one of the central ones) can be resolved unless the excited state decay rate is smaller than the recoil shift. Since this condition is violated for allowed electronic transitions, it is of some interest to look for alternative level schemes in which this structure

can be resolved fully. If the optical transitions are replaced by two-photon, Raman-like transitions between ground state levels, the widths of the various spectral components are determined by ground state relaxation rates, rather than excited state decay rates. As a result, the probe's spectral response should be fully resolvable. Raman processes have taken on added importance in sub-Doppler [8] and sub-recoil [9] cooling, atom focusing [10], atom interferometry [11–14], and as a method for probing Bose condensates [15].

In this article we propose a scheme for pump-probe spectroscopy of an atomic vapor using Raman transitions. This is but one of a class of interactions that can be considered under the general heading of *nonlinear ground state spectroscopy*. The spectral response is found to be similar to that of traditional pump-probe spectroscopy [1]; however, new interference phenomena can modify the spectrum [Sec. III]. The interference phenomena can be interpreted in terms of a dressed atom picture [Sec. IV]. Although part of the motivation for this work is the study of recoil effects, such effects are neglected in this article.

II. EQUATIONS OF MOTION

The atom field geometry is indicated schematically in Fig. 1. Three-level atoms interact with two optical fields, E_1 and E_2 , producing strong coupling between initial and final levels 1 and 2 via an intermediate excited state level e . Field E_1 couples only levels 1 and e , while field E_2 couples only levels 2 and e . In addition, there is a weak probe field E that couples only levels 1 and e . As a consequence, fields E and E_2 can also drive two-photon transitions between levels 1 and 2. Levels 1 and 2 are pumped incoherently at rates Λ_1 and Λ_2 , respectively, and both states decay at rate Γ . The incoherent pumping and decay represent an oversimplified model for atoms entering and leaving the interaction volume.

FIG. 1. Schematic diagram of the atom-field system. Fields E_1 and E drive only the $1-e$ transition and field E_2 only the $2-e$ transition.

$$E(\mathbf{R}, t) = \frac{1}{2} \left[E_1 e^{i(\mathbf{k}_1 \cdot \mathbf{R} - \Omega_1 t)} + E_2 e^{i(\mathbf{k}_2 \cdot \mathbf{R} - \Omega_2 t)} + E e^{i(\mathbf{k} \cdot \mathbf{R} - \Omega t)} \right] + c.c., \quad (1)$$

where Ω_1 , Ω_2 , and Ω are the field frequencies, \mathbf{k}_1 , \mathbf{k}_2 , and \mathbf{k} the field propagation vectors, and $c.c.$ stands for complex conjugate. In an interaction representation, neglecting any decay or incoherent pumping of the ground state levels, the state probability amplitudes obey the equations of motion.

$$i\dot{a}_e = \chi_1 e^{-i\Delta_1 t} a_1 + \chi_2 e^{-i\Delta_2 t} a_2 + \chi e^{-i\Delta t} a_1 - i(\gamma_e/2) a_e, \quad (2a)$$

$$i\dot{a}_1 = \chi_1 e^{i\Delta_1 t} a_e + \chi e^{i\Delta t} a_e, \quad (2b)$$

$$i\dot{a}_2 = \chi_2 e^{i\Delta_2 t} a_e, \quad (2c)$$

where $\chi_j = -d_{ej}E_j/2\hbar$ ($j = 1, 2$) and $\chi = -d_{e1}E/2\hbar$ are Rabi frequencies (assumed to be real and positive), d_{ej} is a dipole moment matrix element, and $\Delta_j = \Omega_j - \omega_{ej}$ and $\Delta = \Omega - \omega_{e1}$ are atom-field detunings. Assuming that the magnitude of the detunings are much larger than γ_e and any Doppler shifts associated with the single photon transitions, it is possible to adiabatically eliminate the excited state amplitude to arrive at the following equations for the ground state amplitudes:

$$i\dot{a}_1 = S_1 a_1 + S(e^{i\delta_1 t} + e^{-i\delta_1 t}) a_1 + g e^{-i\tilde{\delta} t} a_2 + g' e^{-i\delta' t} a_2, \quad (3a)$$

$$i\dot{a}_2 = S_2 a_2 + g e^{i\tilde{\delta} t} a_1 + g' e^{i\delta' t} a_1, \quad (3b)$$

where

$$\tilde{\delta} = \Delta_2 - \Delta_1 = \Omega_2 - \Omega_1 + \omega_{21}; \quad (4a)$$

$$\delta' = \Delta_2 - \Delta = \Omega_2 - \Omega + \omega_{21}; \quad (4b)$$

$$\delta_1 = \Delta - \Delta_1 = \Omega - \Omega_1 = \tilde{\delta} - \delta', \quad (4c)$$

The incident fields are assumed to be nearly copropagating so that all two-photon Doppler shifts can be neglected. In this limit and in the limit of large detuning on each single photon transition, one can consider the atoms to be stationary with regards to their interaction with the external fields. We wish to calculate the linear probe absorption spectrum. The electric field can be written as

are detunings associated with two-quantum processes and

$$g = \chi_1 \chi_2 / \Delta; \quad g' = \chi \chi_2 / \Delta; \\ S_1 = \chi_1^2 / \Delta; \quad S_2 = \chi_2^2 / \Delta; \quad S = \chi \chi_1 / \Delta, \quad (5)$$

are Rabi frequencies or Stark shifts associated with two quantum processes. In writing Eqs. (3), we assumed that $\Delta \approx \Delta_1 \approx \Delta_2$ and $|\Delta| \gg |\tilde{\delta}|, |\delta'|, |\delta_1|$.

It will prove convenient, especially when going over to a dressed atom picture, to introduce a representation in which

$$a_1 = b_1 e^{-i\tilde{\delta} t/2} e^{-i(S_1 + S_2)t/2}; \\ a_2 = b_2 e^{i\tilde{\delta} t/2} e^{-i(S_1 + S_2)t/2}. \quad (6)$$

Combining Eqs. (3) and (6) one finds

$$i\dot{b}_1 = -(\delta/2)b_1 + gb_2 + S(e^{i\delta_1 t} + e^{-i\delta_1 t})b_1 + g'e^{i\delta_1 t}b_2; \quad (7a)$$

$$i\dot{b}_2 = (\delta/2)b_2 + gb_1 + g'e^{-i\delta_1 t}b_1, \quad (7b)$$

where

$$\delta = \tilde{\delta} - (S_1 - S_2). \quad (8)$$

The corresponding equations for density matrix elements $\rho_{11} = |b_1|^2$, $\rho_{22} = |b_2|^2$, $\rho_{12} = b_1 b_2^* = \rho_{21}^*$ are

$$\dot{\rho}_{11} = -ig(\rho_{21} - \rho_{12}) - ig'e^{i\delta_1 t}\rho_{21} + ig'e^{-i\delta_1 t}\rho_{12} - \Gamma\rho_{11} + \Lambda_1; \quad (9a)$$

$$\dot{\rho}_{22} = ig(\rho_{21} - \rho_{12}) + ig'e^{i\delta_1 t}\rho_{21} - ig'e^{-i\delta_1 t}\rho_{12} - \Gamma\rho_{22} + \Lambda_2; \quad (9b)$$

$$\dot{\rho}_{12} = i\delta\rho_{12} - ig(\rho_{22} - \rho_{11}) - ig'e^{i\delta_1 t}(\rho_{22} - \rho_{11}) - iS(e^{i\delta_1 t} + e^{-i\delta_1 t})\rho_{12} - \Gamma\rho_{12}, \quad (9c)$$

where the incoherent pumping and decay terms have been introduced. It is important to note that, in this representation, the frequency appearing in the g' terms is $\delta_1 = \delta' - \tilde{\delta} = \Omega - \Omega_1$. In other words, the effective field frequency associated with field E_2 in this representation

is Ω_1 rather than Ω_2 .

It follows from the Maxwell-Bloch equations that the probe absorption coefficient, α , and index change, Δn , are given by

$$\alpha = \frac{kN d_{1e}^2}{2\hbar\epsilon_0} \text{Im} \left(\frac{\rho'_{1e}}{\chi} \right); \quad (10a)$$

$$\Delta n = -\frac{N d_{1e}^2}{2\hbar\epsilon_0} \text{Re} \left(\frac{\rho'_{1e}}{\chi} \right), \quad (10b)$$

where N is the atomic density,

$$\rho'_{1e} \approx \frac{1}{\Delta} \left[\chi \rho_{11}^{(0)} + \chi_1 \rho_{11}^+ + \chi_2 \rho_{12}^+ \right], \quad (11)$$

and $\rho_{11}^{(0)}$, ρ_{11}^+ , and ρ_{12}^+ are coefficients that appear in the solution of Eqs. (9) (to first order in χ) written in the form:

$$\rho_{jj'} = \rho_{jj'}^{(0)} + \rho_{jj'}^+ e^{i\delta_1 t} + \rho_{jj'}^- e^{-i\delta_1 t}; \quad j, j' = 1, 2 \quad (12)$$

The first and third terms in Eq. (11) are analogous to the terms that appear in conventional theories of pump-probe spectroscopy, but the second term is new and leads to qualitatively new features in the probe absorption spectrum.

An expression for ρ'_{1e} is given in Appendix A. The absorption coefficient is plotted in Figs. 2(a)-(c) for several values of δ/g , and

$$\eta = \sqrt{\chi_1/\chi_2}. \quad (13)$$

If $\eta \ll 1$, the two-quantum probe absorption spectrum has the same structure as the probe absorption spectrum involving single quantum transitions. The situation changes if $\eta \gtrsim 1$. For example, aside from an interchange of absorption and gain components as a function of δ , the probe spectrum for single quantum transitions depends only on the magnitude of the pump field detuning. This is clearly *not* the case for two-quantum transitions, as is evident from Fig. 2(a) drawn for $\eta = 1$, $\Gamma/g = 0.1$, $\delta/g = \pm 1$. Probe absorption and gain *are* interchanged when δ changes sign, but the ratio of the amplitude of the absorption to gain peak *changes* when δ changes sign. There is another subtle difference present in these spectra. The sense of the central dispersive component is opposite to that for single quantum transitions. With decreasing η , the sense of the central component would reverse, as the spectrum reverts to the same structure found in pump-probe spectroscopy of single quantum transitions. The probe response also depends on the sign of Δ (through $g = \chi_1 \chi_2 / \Delta$); this feature follows from the dependence of the spectrum on the sign of δ and the relationship

$$\rho'_{1e}(-\delta, -\Delta, -\delta_1) = -\rho'_{1e}(\delta, \Delta, \delta_1)^*, \quad (14)$$

which can be derived using Eqs. (A3)-(A7) of Appendix A. It is also possible for the components centered at positive or negative δ_1 to vanish (in the secular approximation) for certain values of η , as can be seen in Fig. 2(b).

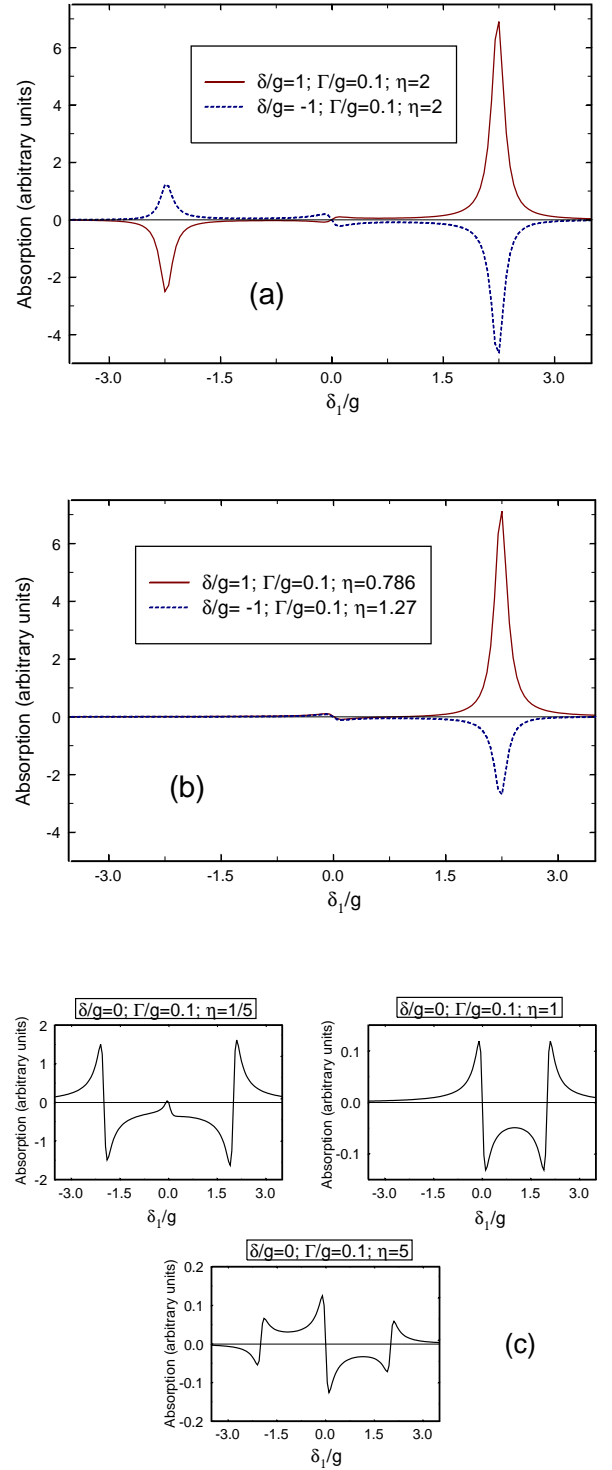


FIG. 2. Probe field absorption in arbitrary units. Positive ordinate values correspond to probe absorption and negative values to probe gain.

The case of $\delta/g = 0$ is shown in Fig. 2(c) for $\eta = 1/5, 1, 5$, and $\Delta > 0$. If $\eta = 1/5$, the spectrum is similar to that found for single quantum transitions [1]. For $\eta = 1$, the spectral component at negative δ_1 is found to vanish. When $\eta \gtrsim 1$, there is a dispersive-like structure centered at $\delta_1 = 0$ that is not found in the pump-probe

FIG. 3. Dressed-state energy level diagram. In the interaction representation adopted in the text, the frequency of field E_2 must be set equal to Ω_1 in calculating resonance conditions. For $(\Lambda_A - \Lambda_B) > 0$, solid arrows correspond to probe absorption centered at $\delta_1 = \omega_{BA}$ and dashed arrows correspond to probe gain centered at $\delta_1 = -\omega_{BA}$.

It is now possible to use the energy level diagram (Fig. 3) to read directly the probe absorption spectrum. The probe field is absorbed (or amplified) via two quantum transitions between states $|A\rangle$ and $|B\rangle$. The two quantum transitions involve one photon from the probe field and one photon from *either* field E_1 or E_2 , since all of these fields couple states $|A\rangle$ and $|B\rangle$ to state $|e\rangle$. It is important to remember that the effective field frequency of field E_2 is equal to Ω_1 in this interaction representation. Fields E_1 and E couple state $|e\rangle$ to the components of states $|A\rangle$ and $|B\rangle$ involving state $|1\rangle$, while field E_2 couples state $|e\rangle$ to the components of states $|A\rangle$ and $|B\rangle$ involving state $|2\rangle$. For example the matrix element for the two-quantum process from state $|A\rangle$ to $|B\rangle$ involving absorption of a probe photon and emission of a field E_2 photon is

$$\frac{-i\chi}{-i\Delta} \cos(\theta) \frac{(-i\chi_2)}{\Gamma - i(\delta_1 - \omega_{BA})} \cos(\theta),$$

while that for absorption of a probe photon and emission of a field E_1 photon is

$$\frac{-i\chi}{-i\Delta} \cos(\theta) \frac{(-i\chi_1)}{\Gamma - i(\delta_1 - \omega_{BA})} \psi \sin(\theta).$$

These two processes add coherently, such that probe absorption via transitions from state $|A\rangle$ to $|B\rangle$ is proportional to the sum of these two matrix elements squared, multiplied by the population difference $(\rho_{AA}^{(0)} - \rho_{BB}^{(0)})$. In other words, the probe absorption at $\delta_1 = \omega_{BA}$ is proportional to a quantity C_+ given by

$$C_+ = (g/\Delta\Gamma) [(\Lambda_1 - \Lambda_2)/\Gamma] \cos(2\theta) \left(\psi \eta \sin(\theta) \cos(\theta) + \frac{1}{\eta} \cos^2(\theta) \right)^2. \quad (21)$$

Similarly, probe gain via transitions from state $|A\rangle$ to $|B\rangle$ at $\delta_1 = -\omega_{BA}$ is proportional to

$$C_- = (g/\Delta\Gamma) [(\Lambda_1 - \Lambda_2)/\Gamma] \cos(2\theta) \left(\psi \eta \sin(\theta) \cos(\theta) - \frac{1}{\eta} \sin^2(\theta) \right)^2. \quad (22)$$

A formal derivation of these results is given in Appendix B.

For the sake of definiteness, let us take $(\Lambda_1 - \Lambda_2) > 0$; then C_+ corresponds to absorption for $\delta > 0$ and to gain for $\delta < 0$, while C_- corresponds to gain for $\delta > 0$ and to absorption for $\delta < 0$. Note that the component centered at $\delta_1 = -\omega_{BA}$ vanishes if $\Delta > 0$ and $\tan(\theta) = \eta^2$, while that at $\delta_1 = \omega_{BA}$ vanishes if $\Delta < 0$ and $\tan(\theta) = \eta^{-2}$. The values of $A_{\pm} = \pm C_{\pm} [\Gamma^2 \Delta / |g| (\Lambda_1 - \Lambda_2)]$ are plotted in Fig. 4 as a function of δ/g for $\Delta > 0$ and $\eta = 1, 2$. For $\Delta < 0$, one can use the relationship $A_{\pm}(-\Delta, -\delta) = A_{\mp}(\Delta, \delta)$.

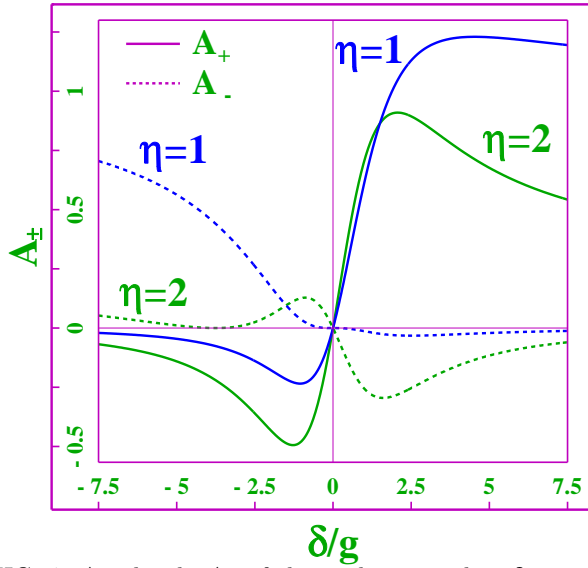


FIG. 4. Amplitude A_+ of the peak centered at $\delta_1 = \omega_{BA}$ and amplitude A_- of the peak centered at $\delta_1 = -\omega_{BA}$, for $\Delta > 0$. Positive values of A_{\pm} correspond to absorption and negative values to gain.

The probe absorption vanishes in the secular approximation (19) when $\delta = 0$, since, in this case, $\theta = \frac{\pi}{4}$ and the populations of the dressed states are equal. The lowest order dressed atom approach is not useful in this limit. Typical spectra are shown in Fig. 2(c) and were discussed in Sec. III.

IV. CONCLUSION

The probe absorption spectrum has been calculated for two-quantum transitions between levels that are simultaneously driven by a two-quantum pump field of arbitrary intensity. In addition to features found in conventional pump-probe spectroscopy of single quantum transitions, new features have been found that can be identified with interference phenomena. Both Doppler and recoil effects were neglected in our treatment. For nearly copropagating fields, effects arising from these processes are negligible. Doppler shifts can be accounted for by the replacements $\delta_1 \rightarrow \delta_1 + (\mathbf{k}_1 - \mathbf{k}) \cdot \mathbf{v}$, $\delta_1 - \tilde{\delta} \rightarrow \delta_1 - \tilde{\delta} + (\mathbf{k}_2 - \mathbf{k}) \cdot \mathbf{v}$, and $\delta_1 + \tilde{\delta} \rightarrow \delta_1 + \tilde{\delta} + (2\mathbf{k}_1 - \mathbf{k}_2 - \mathbf{k}) \cdot \mathbf{v}$ in the equations in the Appendix.

The dependence of the interference effect of the signs of Δ and $\tilde{\delta}$ can be understood in the bare atom picture in a perturbative limit. A schematic representation of the probability amplitude leading to probe absorption at $\delta_1 = \tilde{\delta}$ is shown in Fig. 5(a). Each arrow represents an interaction with one of the fields. The two contributions to the final state amplitude add coherently. Putting in the appropriate energy denominators, one finds that the absorption varies as

$$A = \left| \frac{i^2 \chi \chi_2^*}{(\gamma_e/2 - i\Delta) [\Gamma - i(\Delta - \Delta_2)]} + \frac{i^4 \chi \chi_2^* |\chi_1|^2}{(\gamma_e/2 - i\Delta) [\Gamma - i(\Delta - \Delta_1)] (\gamma_e/2 - i\Delta) [\Gamma - i(\Delta - \Delta_2)]} \right|^2. \quad (23)$$

For $|\tilde{\delta}| \gg \Gamma$, and $|\Delta| \gg \gamma_e$, this equation reduces to

$$A = \left| \frac{\chi \chi_2^*}{\Delta} \right|^2 \frac{1}{\Gamma^2 + (\delta_1 - \tilde{\delta})^2} \left| 1 + \frac{|g| \eta^2 \psi}{\tilde{\delta}} \right|, \quad (24)$$

which shows the dependence on the signs of Δ ($\psi = |\Delta|/\Delta$) and $\tilde{\delta}$. A similar calculation for the emission com-

ponent represented schematically in Fig. 5(b) leads to

$$G = \left| \frac{\chi^* \chi_1^2 \chi_2^*}{\Delta^2 \tilde{\delta}} \right|^2 \frac{1}{\Gamma^2 + (\delta_1 + \tilde{\delta})^2} \left| 1 - \frac{|g| \eta^{-2} \psi}{\tilde{\delta}} \right|. \quad (25)$$

New effects will arise if the fields are not copropagating and the active medium is a subrecoil cooled atomic

FIG. 5. Schematic representation of the $1 \rightarrow 2$ transition probability leading to probe absorption or probe gain in lowest order perturbation theory in the bare basis. The thin arrow represents the probe field, the broad filled arrows field E_1 , and the broad open arrows field E_2 . (a) absorption, (b) gain. Terms involving the sequential absorption and emission of the *same* field have been neglected, since such terms result only in Stark shifts of levels 1 and 2. The diagrams are drawn for $\tilde{\delta} > 0$; if $\tilde{\delta} < 0$, the roles of absorption and gain would be interchanged.

vapor, a highly collimated atomic beam, or a BEC. As for single quantum transitions [7], each component of the spectrum undergoes recoil splitting. Since the center-of-mass momentum states differ for two-quantum processes involving fields E_1 and E from those involving fields E_2 and E , one might expect the spectrum consists of eight absorption and eight emission components rather than the four absorption and four emission components found for single quantum transitions; however this does not appear to be the case. Instead, each component results from a coherent superposition of two quantum processes

involving fields (E_1, E) and (E_2, E) .

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APPENDIX A: BARE STATE CALCULATIONS

Substituting Eqs. (12) into Eqs. (9), one finds to zeroth order in the probe field that

$$w^{(0)} = \rho_{22}^{(0)} - \rho_{11}^{(0)} = \frac{(\Lambda_2 - \Lambda_1)}{\Gamma} \frac{\Gamma^2 + \delta^2}{\Gamma^2 + \delta^2 + 4|g|^2}; \quad (\text{A1a})$$

$$\rho_{11}^{(0)} = \frac{1}{2} \left[\frac{(\Lambda_2 + \Lambda_1)}{\Gamma} - w^{(0)} \right]; \quad (\text{A1b})$$

$$\rho_{12}^{(0)} = \frac{-ig}{\Gamma - i\delta} w^{(0)} = \left(\rho_{21}^{(0)} \right)^*, \quad (\text{A1c})$$

and that, to first order in the probe field, $w^+ = \rho_{22}^+ - \rho_{11}^+$, ρ_{12}^+ , ρ_{21}^+ , and $m^+ = \rho_{22}^+ + \rho_{11}^+$ satisfy

$$m^+ = 0; \quad (\text{A2a})$$

$$(\Gamma + i\delta_1) w^+ - 2ig^* \rho_{21}^+ + 2ig \rho_{12}^+ = 2ig' \rho_{21}^{(0)}; \quad (\text{A2b})$$

$$[\Gamma + i(\delta_1 - \delta)] \rho_{12}^+ + ig^* w^+ = -ig'^* w^{(0)} - iS \rho_{12}^{(0)}; \quad (\text{A2c})$$

$$[\Gamma + i(\delta_1 + \delta)] \rho_{21}^+ - ig w^+ = iS \rho_{21}^{(0)}. \quad (\text{A2d})$$

Equation (11) can be rewritten as

$$\rho'_{1e} \approx (\chi^*/\Delta) \left[\rho_{11}^{(0)} - \frac{1}{2} y_0 + y_{12} \right], \quad (\text{A3})$$

where

$$y_0 = (\chi_1/\chi)^* w^+; \quad (\text{A4a})$$

$$y_{12} = (\chi_2/\chi)^* \rho_{12}^+; \quad (\text{A4b})$$

$$y_{21} = \left(\frac{\chi_2 \chi_1^*}{\chi^* \chi_1} \right) \rho_{21}^+. \quad (\text{A4c})$$

In Eqs. (A1-A4) we have allowed the Rabi frequencies to be complex.

The quantities y_0 , y_{12} , and y_{21} satisfy the coupled equations:

$$[\Gamma + i(\delta_1 - \delta)] y_{12} + i|g| \psi \eta^{-2} y_0 = a; \quad (\text{A5a})$$

$$(\Gamma + i\delta_1) y_0 - 2i|g| \psi \eta^2 y_{21} + 2i|g| \psi \eta^2 y_{12} = b; \quad (\text{A5b})$$

$$[\Gamma + i(\delta_1 + \delta)] y_{21} - i|g| \psi \eta^{-2} y_0 = c, \quad (\text{A5c})$$

where

$$b = \frac{-2|g|^2}{\Gamma + i\delta} w^{(0)} = 2c \quad (\text{A6a})$$

$$a = -i|g| \psi \eta^{-2} w^{(0)} - \frac{|g|^2}{\Gamma - i\delta} w^{(0)}. \quad (\text{A6b})$$

and $\psi = (|\Delta|/\Delta)$. Note that the equations do not depend on the phase of the various Rabi frequencies, but *do* depend on the sign of Δ . Explicit solutions for y_0 and y_{12} are:

$$y_0 = -i \frac{2a |g| \psi \eta^2 (\delta + \delta_1 - i\Gamma) + b [\delta^2 - (\delta_1 - i\Gamma)^2] + 2c |g| \psi \eta^2 (\delta - \delta_1 + i\Gamma)}{(\delta_1 - i\Gamma) (\delta^2 - \delta_1^2 + 2i\Gamma\delta_1 + \Gamma^2 + 4|g|^2)}; \quad (\text{A7a})$$

$$y_{12} = i \frac{a [\delta_1^2 + \delta (\delta_1 - i\Gamma) - 2i\Gamma\delta_1 - \Gamma^2 - 2|g|^2] - b\eta^{-2} |g| \psi (\delta + \delta_1 - i\Gamma) - 2c |g|^2}{(\delta_1 - i\Gamma) (\delta^2 - \delta_1^2 + 2i\Gamma\delta_1 + \Gamma^2 + 4|g|^2)}. \quad (\text{A7b})$$

The line shape is totally non-secular when $\delta = 0$. In the limit that $\Delta > 0$, $|g| \gg \Gamma$, and $\Gamma \ll \eta^2$, one finds that the absorption coefficient α for $\delta_1 \approx 0$ is

$$\alpha \sim -\frac{1}{4} \left(\frac{kN d_{1e}^2}{2\hbar\epsilon_0\Delta} \right) \frac{\Lambda_1 - \Lambda_2}{\Gamma} \frac{\delta_1\Gamma}{(\delta_1^2 + \Gamma^2)}, \quad (\text{A8a})$$

that for $\delta_1 \approx 2|g|$ is

$$\alpha \sim \frac{1}{8} \left(\frac{kN d_{1e}^2}{2\hbar\epsilon_0\Delta} \right) \frac{\Lambda_1 - \Lambda_2}{\Gamma} \frac{(\delta_1 - 2|g|)\Gamma}{[(\delta_1 - 2|g|)^2 + \Gamma^2]} (1 + \eta^{-2}), \quad (\text{A8b})$$

and that for $\delta_1 \approx -2|g|$ is

$$\alpha \sim \frac{1}{8} \left(\frac{kN d_{1e}^2}{2\hbar\epsilon_0\Delta} \right) \frac{\Lambda_1 - \Lambda_2}{\Gamma} \frac{(\delta_1 + 2|g|)\Gamma}{[(\delta_1 + 2|g|)^2 + \Gamma^2]} (1 - \eta^{-2}). \quad (\text{A8c})$$

Note that the component at $\delta_1 = -2|g|$ vanishes if $\eta = 1$. For $\Delta < 0$, one can use Eq. (14).

APPENDIX B: DRESSED-STATE CALCULATIONS

Equation (7) can be written in the form

$$i\hbar \dot{\mathbf{b}} = (\mathbf{V} + \mathbf{V}_p) \mathbf{b}, \quad (\text{B1})$$

where

$$\mathbf{V} = \hbar \begin{pmatrix} -\delta/2 & g^* \\ g & \delta/2 \end{pmatrix}, \quad (\text{B2})$$

$$\mathbf{V}_p = \hbar \begin{pmatrix} S e^{i\delta_1 t} + S^* e^{-i\delta_1 t} & g'^* e^{i\delta_1 t} \\ g' e^{-i\delta_1 t} & 0 \end{pmatrix}, \quad (\text{B3})$$

$$g = \frac{\chi_1 \chi_2^*}{\Delta}; \quad g' = \frac{\chi \chi_2^*}{\Delta}; \quad S = \frac{\chi^* \chi_1}{\Delta}, \quad (\text{B4})$$

and we have allowed for complex Rabi frequencies,

$$\left(\frac{d}{dt} + \Gamma \right) \rho_d^+ = i \begin{pmatrix} 0 & (\omega_{BA} - \delta_1) \rho_{AB}^+ \\ -(\omega_{BA} + \delta_1) \rho_{BA}^+ & 0 \end{pmatrix} + \frac{1}{i\hbar} [\mathbf{V}_{pd}, \rho_d^{(0)}], \quad (\text{B14})$$

where

$$\chi_1 = |\chi_1| e^{i\phi_1}, \chi_2 = |\chi_2| e^{i\phi_2}, \chi = |\chi| e^{i\phi}. \quad (\text{B5})$$

If one introduces semi-classical dressed states via the transformation

$$\mathbf{b}_d = \mathbf{T}_c \mathbf{b}, \quad (\text{B6})$$

where

$$\mathbf{b}_d = \begin{pmatrix} A \\ B \end{pmatrix}, \quad (\text{B7})$$

$$\mathbf{T}_c = \begin{pmatrix} \cos(\theta) e^{i\phi_d/2} & -e^{-i\phi_d/2} \sin(\theta) \\ e^{i\phi_d/2} \sin(\theta) & e^{-i\phi_d/2} \cos(\theta) \end{pmatrix}, \quad (\text{B8})$$

and

$$\phi_d = \phi_1 - \phi_2 + \frac{\pi}{2}(1 - \psi) \quad (\text{B9})$$

(recall that $\psi = |\Delta|/\Delta$), then the dressed-state Hamiltonian is given by

$$\mathbf{V}_d = \hbar \begin{pmatrix} -\omega_{BA}/2 & 0 \\ 0 & \omega_{BA}/2 \end{pmatrix} + \mathbf{T}_c \mathbf{V}_p \mathbf{T}_c^\dagger. \quad (\text{B10})$$

The dressed state density matrix,

$$\rho_d = \begin{pmatrix} \rho_{AA} & \rho_{AB} \\ \rho_{BA} & \rho_{BB} \end{pmatrix} \quad (\text{B11})$$

evolves as

$$\left(\frac{d}{dt} + \Gamma \right) \rho_d \approx \frac{1}{i\hbar} [\mathbf{V}_d, \rho_d] + \begin{pmatrix} \Lambda_A & 0 \\ 0 & \Lambda_B \end{pmatrix}, \quad (\text{B12})$$

Off-diagonal terms have been neglected in the matrix representing the incoherent pumping, since they give rise to terms of order $\Gamma/\omega_{BA} \ll 1$ (secular approximation).

The dressed state density matrix is expanded as

$$\rho_d = \rho_d^{(0)} + \rho_d^+ e^{i\delta_1 t} + \rho_d^- e^{-i\delta_1 t}, \quad (\text{B13})$$

and it is found from Eqs. (B1)-(B3), (B6)-(B13) that ρ_d^+ obeys the equation of motion

$$\mathbf{V}_{pd} = \hbar \begin{pmatrix} \cos(\theta) [S \cos(\theta) - g'^* e^{i\phi_d} \sin(\theta)]; & \cos(\theta) [S \sin(\theta) + g'^* \cos(\theta) e^{i\phi_d}] \\ \sin(\theta) [-g'^* \sin(\theta) e^{i\phi_d} + S \cos(\theta)]; & \sin(\theta) [S \sin(\theta) + g'^* \cos(\theta) e^{i\phi_d}] \end{pmatrix} \quad (\text{B15})$$

In the secular approximation, the steady state solution of Eq. (B14) is

$$\rho_d^+ = \begin{pmatrix} 0 & \rho_{AB}^+ \\ \rho_{BA}^+ & 0 \end{pmatrix}, \quad (\text{B16})$$

where

$$\rho_{AB}^+ = i \cos(\theta) [S \sin(\theta) + g'^* \cos(\theta) e^{i\phi_d}] (\rho_{AA}^{(0)} - \rho_{BB}^{(0)}) / (\Gamma + i(\delta_1 - \omega_{BA})), \quad (\text{B17a})$$

$$\rho_{BA}^+ = -i \sin(\theta) [-g'^* \sin(\theta) e^{i\phi_d} + S \cos(\theta)] (\rho_{AA}^{(0)} - \rho_{BB}^{(0)}) / (\Gamma + i(\delta_1 + \omega_{BA})). \quad (\text{B17b})$$

The coherence ρ'_{1e} needed in Eq. (10) for the absorption coefficient and index change is given by

$$\rho'_{1e} \approx (1/\Delta) [\chi^* \rho_{11}^{(0)} + \chi_1^* \rho_{11}^+ + \chi_2^* \rho_{12}^+]. \quad (\text{B18})$$

The first term can be evaluated using Eq. (A1b) for $\rho_{11}^{(0)}$; it contributes to the index change, but not the absorption. For the remaining terms, one rewrites ρ_{11}^+ and ρ_{12}^+ in the dressed basis using Eqs. (B6),(B8),(B11), and uses Eq. (B5) to extract all the phase factors to arrive at

$$\rho'_{1e} \approx (\chi^* / |\Delta|) [\psi \rho_{11}^{(0)} + f_+ + f_-] \quad (\text{B19})$$

where

$$f_+ = \frac{i|g|}{[\Gamma + i(\delta_1 - \omega_{BA})]} \cos(2\theta) \frac{(\Lambda_1 - \Lambda_2)}{\Gamma} \cos^2(\theta) \left(\psi \eta \sin(\theta) + \frac{1}{\eta} \cos(\theta) \right)^2, \quad (\text{B20a})$$

$$f_- = -\frac{i|g|}{[\Gamma + i(\delta_1 + \omega_{BA})]} \cos(2\theta) \frac{(\Lambda_1 - \Lambda_2)}{\Gamma} \sin^2(\theta) \left(\psi \eta \cos(\theta) - \frac{1}{\eta} \sin(\theta) \right)^2. \quad (\text{B20b})$$

Note that the approach and results of Sec. III are unchanged if one uses complex dressed states defined by

$$\begin{pmatrix} |A\rangle \\ |B\rangle \end{pmatrix} = \mathbf{T}_c^* \begin{pmatrix} |1\rangle \\ |2\rangle \end{pmatrix}. \quad (\text{B21})$$

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